NASA TECHNICAL TRANSLATION

NASA TT F-11,470

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Translation of "O vzaimodeystvii atomov s poverkhnost'yu tverdogo tela"
Zhurnal Prikladnoy Mekhaniki i Tekhnicheskoy Fiziki,
No. 4, July-Aug. 1967, pp. 113-117

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON, D.C., 20546 JANUARY 1968

NASA TT F-11,470

ON THE INTERACTION OF ATOMS WITH THE SURFACE OF A SOLID

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ABSTRACT: Computer study of the interaction of atoms with a solid body at energies on the order of 10 eV. The procedure is based on the simultaneous solution of the system of classical equations of motion of the atoms (which form the solid body) and the bombarding particle for a given pair-interaction law. A system of dimensionless parameters characterizing such interaction processes is obtained, and typical results of the processes (such as reflection, adsorption, and absorption by the lattice of the incident particle) are demonstrated. The interaction of an atom with an ideal crystal with an Einstein lattice is examined for various parameters and lattice types. Estimates are obtained of the effect of the type of interaction law on the statistically averaged interaction characteristics (energy accommodation coefficient). A method for averaging the interaction process of atoms with an atomically smooth polycrystalline surface. V. P.

Some results of a computer study of the process of interaction of atoms with a solid surface at energies of ~10 eV are presented. The method is based on the simultaneous solution of a system of the classical equations for the motion of the atoms of the solid, and of the bombarding particle, given some law for pair interaction. The 6-12 Lennard-Jones potential is used as this law in most calculations. /113*

A system of dimensionless parameters is obtained, which characterizes the processes being considered, and typical results of the interaction event are shown: reflection, capture by the surface (adsorption), absorption of the incident particle by the lattice. The interaction of an atom with the surface of an ideal crystal, forming an Einstein lattice, is examined for different parameters and lattice types, and the effects of the potential form and parameters on the statistically averaged characteristics of the interaction are evaluated (energy accommodation coefficient). A method is described for averaging the interaction characteristics that makes it possible to simulate the collision process of atoms with an atomically smooth polycrystalline surface.

1. Method. System of controlling parameters. The experimental difficulties of producing intense atomic beams with energies of ~10 eV and, also, the complicated problem of identifying the conditions at the interaction surface in

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*Numbers in the margin indicate pagination in the foreign text.

different experimental apparatus lead to a wide dispersion in the value of the characteristics of the interaction process of atomic beams with a solid surface.

Work, devoted to a theoretical examination of this process, is based primarily on a simplification of the lattice model, which is replaced by a one-dimensional array or two-dimensional lattice of atoms. Moreover, a simplified pair interaction law is also frequently assumed. All this enables one to consider the results of such investigations as being qualitative in nature. Only in [1, 2] is an attempt made to examine the problem in a more rigorous fashion (three-dimensional lattice, an interaction potential that approximates reality) and with a minimum number of simplifying assumptions. This treatment enables one to obtain data suitable for comparison to the results of experimental investigations, and also to determine the degree of influence of different controlling parameters. No less important is the possibility of defining functions for the energy and directional distribution of reflected particles.

To this end, an array of atoms in a solid with a regular crystalline structure of one type or another is considered: simple cubic (SC), face-centered cubic (FCC) and body-centered cubic (BCC). It is assumed that for a bombarding particle energy of ~10 eV the thermal vibrations of the lattice atoms can be neglected. The assumption that the probability of two or more particles simultaneously entering a typical region is negligibly small, for the actual flux densities of atomic beams, is also obvious.

Let the interaction of an incident particle with each of the atoms of the array be determined by the potential V(r), and let the atoms themselves form an Einstein crystal. The latter is true only if the interaction time is less than the period of the natural vibrations of the lattice atoms or, what is sufficient, the average velocity of the incident atom is greater than the propagation velocity of elastic vibrations in the solid.

After selection of a crystal lattice and its orientation with respect to the free surface (the 100 face in the present calculations), the interaction process is determined by the following parameters: the initial energy of the incident particle, E_0; the mass of the incident particle, m_1; the mass of the lattice atom, m_2; the parameters of the 6-12 Lennard-Jones potential, ϵ and σ; the crystal lattice spacing, d; the stiffness coefficient $\varkappa$ of the spring with which a lattice atom is bound to the normal equilibrium position; the angle to the normal, φ, and the azimuth, ψ, of the initial velocity direction of the incident particle; the rectangular coordinates of the target point (TP) on the crystal surface, x_t, y_t.

Thus, the energy accommodation coefficient α will depend on eight dimensionless parameters

$$\mu = \frac{m_1}{m_2}, \quad w = \left(\frac{2E_0}{\mu\sigma^2\varkappa}\right)^{1/2}, \quad \varphi, \quad s = \frac{24\varepsilon}{\sigma^2\varkappa}, \quad a = \frac{d}{\sigma}, \quad \xi = \frac{x_t}{\sigma}, \quad \zeta = \frac{y_t}{\sigma}, \quad \psi$$

The accommodation coefficients, averaged over the last three parameters, will obviously be of practical interest.

Let us select a rectangular coordinate system with its origin at one of the surface atoms of the lattice in such a manner that the plane z = 0 coincides with the free surface of the crystal and the z-axis is directed toward empty space. /114
We will consider σ as a linear scale, $(m_2/\varkappa)^{1/2}$ as a time scale. Then the system of equations describing the motion of the incident atom and the crystal atoms will have the form

$$\frac{d^2R_0}{d\tau^2} = -\frac{1}{\mu} s \sum_{l,m,n} f(|R_0 - R_{lmn}|) \frac{R_0 - R_{lmn}}{|R_1 - R_{lmn}|}$$
$$\frac{d^2R_{lmn}}{d\tau^2} = sf(|R_0 - R_{lmn}|) \frac{R_0 - R_{lmn}}{|R_0 - R_{lmn}|} + L_{lmn} \qquad (1.1)$$
$$f(q) = q^{-7} - 2q^{-13}$$

Here R_0 is the radius vector of the incident particle; R_{lmn} is the radius vector of the lattice atom with the number l, m, n; L_{lmn} is the force acting on the lattice atom with number l, m, n from the crystal side; τ is the time.

Because the interaction potential rapidly approaches zero as the distance increases, not all the crystal atoms participate in the interaction process, but only those that lie within some neighborhood of the target atom. For an Einstein crystal the number of atoms, whose effect should be taken into account completely, is determined by the type of interaction potential. It will be shown below that for the 6-12 Lennard-Jones potential and not too large values of φ, ignoring all atoms located at a distance of $\geq 3\sigma$ from the target atom does not lead to any significant errors in the calculation of the accommodation coefficient.

The initial conditions (at $\tau = 0$) for the solution of the system of equations (1.1) were defined in the following manner:

$$R_{0x} = \xi - \eta_0 \operatorname{tg}\varphi \cos\psi$$
$$R_{0y} = \zeta - \eta_0 \operatorname{tg}\varphi \sin\psi$$
$$R_{0z} = \eta_0 = z_0/\sigma$$
$$dR_{0x}/d\tau = w \sin\varphi \cos\psi \qquad (1.2)$$
$$dR_{0y}/d\tau = w \sin\varphi \sin\psi$$
$$dR_{0z}/d\tau = -w \cos\varphi$$
$$dR_{lmn}/d\tau = 0$$

At the initial instant of time the values of the vectors R_{lmn} correspond to the equilibrium positions in the chosen crystal type.

Based on what has been said above, we set $\eta_0 = 3$. Test calculations, as η_0 was increased, demonstrated the validity of this assumption.

Integration of Eqs. (1.1) for the initial conditions (1.2) was done by the Runge-Kutta method on a BESM-2M computer with variable integration interval. A test of accuracy was made in terms of the total energy.

2. Possible results of the interaction process. The interaction process can lead to three fundamentally different results, each of which actually occurs in the analyses performed.

1. Reflection. After interaction the particle leaves the zone of influence of the crystal and proceeds with uniform rectilinear motion. In this case the integration was terminated when the particle had moved a distance η_0 from the solid. The accommodation coefficient was computed from the formula

$$\alpha = 1 - E_f/E_0$$

where E_f is the particle energy after the interaction.

2. Capture. This is observed for a low initial incident particle energy. /115
As a result of the interaction the particle loses its normal component of velocity and, remaining within the solid, is either trapped in the potential well of one of the atoms or continues to move along an equipotential surface, not escaping from the crystal. The accommodation coefficient for such trajectories is assumed equal to 1.

Theoretical works, based on a one-dimensional crystal model, give the dependence of the ratio of the threshold capture energy to the depth of the potential well on μ and the type of coupling of the incident atom with the first atom of the lattice. In the symbols employed here, this ratio is equal to $12\mu w^2/s$. Continuing, let us point out that in the present analyses capture is observed at energies that are twenty times greater than those which are indicated in these works. This discrepancy is apparently associated with the three-dimensional model used by us.

3. Absorption. This is observed with large separations between the atoms in the crystal and high incident atom energies. As a result of the interaction the particle enters the solid and either sticks in the selected array of atoms or passes completely through it, retaining the momentum within the solid. The accommodation coefficient in this case was also assumed equal to unity. Examples of typical trajectories for the different interaction cases are shown in Fig. 1, a-d. It must be noted that for clarity only planar trajectories are illustrated, for which the initial velocity vector of the incident particle lies in the symmetry plane of the crystal.

Shown in Fig. 2 is the time dependence of the incident particle energy E for the trajectory shown in Fig. 1, a. As seen from the graph, the interaction time for this case is equal to approximately 2.5 and is less than the period of the normal vibrations of a crystal atom (2π in our variables). This confirms the validity of the assumption concerning the insignificance of the elastic wave propagation process in the crystal in the calculation of the accommodation coefficient (in our range of parameters).

3. Accuracy. To evaluate the error, associated with the finiteness of the selected array of atoms, calculations were made of the accommodation coefficient for different array sizes. The effect of the array size on the results of individual trajectory calculations and on the average parameters (concerning the

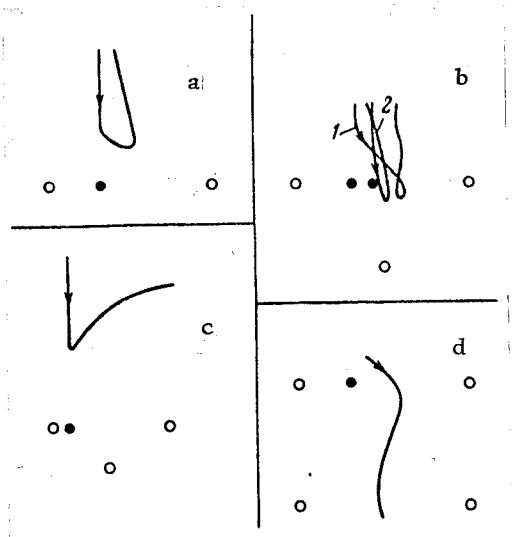

Figure 1. Examples of Typical Trajectories (●—Target Point, o—Lattice Atoms): a) Reflection (SC, $a = 1$, $w = 1$, $s = 0.01$, $\mu = 0.5$, $\xi = \zeta = 0.33$, $\varphi = 0$); b) Reflection (FCC, $a = 1.7$, $w = 1$, $s = 0.01$, $\mu = 0.5$, $\varphi = 0$, $\zeta = 0$, $\xi = 0.56$ (1), $\xi = 0.79$ (2)) c) Capture (BCC, $a = 0.8$, $w = 0.3$, $s = 0.01$, $\mu = 0.5$, $\varphi = 0$, $\xi = \zeta = 0.095$) d) Adsorption (SC, $a = 1.2$, $w = 1$, $s = 0.01$, $\mu = 0.5$, $\varphi = 0$, $\xi = \zeta = 0.40$).

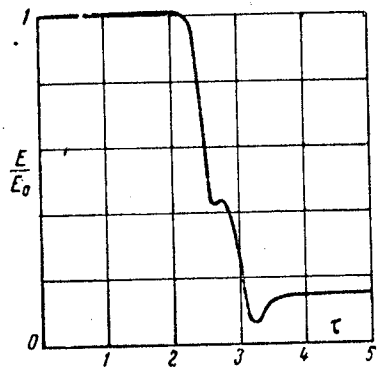

Figure 2. The Time Dependence of the Particle Energy for the Trajectory Illustrated in Fig. 1, a.

averaging, see below) was investigated for the following case: w = 1, $\mu = 0.5$, s = 0.01, a = 1. The results are presented in Table 1.

An array comprising 59 atoms was used in all subsequent calculations. The error in the magnitude of α, contributed by the neglected remaining portion of the crystal, as seen from Table 1, always has a minus sign (i.e., it leads to a decrease in the value of α) and amounts to $\sim 5 \times 10^{-4}$. This choice of array leads to the solving of a system of 360 first order differential equations.

The average of the accommodation coefficient over the target points and φ was done with the quadratic Gauss formulas. For different particle incidence angles the character of the dependence of α on the target point and azimuth is altered—at normal incidence it is

TABLE 1. NUMBER OF ATOMS IN ARRAY

No. of atoms in array	α ($\xi = \zeta = 0$)			$\langle\alpha\rangle$
	BCC	SC	FCC	BCC
34			0.8876	
39	0.9016	0.8971		
50			0.8903	
51	0.9019	0.8972		0.7970
59	0.9022	0.8973	0.8909	0.7975

independent of azimuth and has its strongest dependence on the target point while, on the contrary, at high angles of incidence the dependence on target point flattens out and the azimuth dependence takes on a very complex character. Therefore the choice of methods for averaging over the target points and the evaluation of the errors, produced by this averaging, were done at $\varphi = 0$. A comparison of the results of the calculation of the average accommodation coefficient over 6 ($\langle\alpha\rangle_6$) and over 10 ($\langle\alpha\rangle_{10}$) trajectories, made for all types of crystal lattices, showed that $\langle\alpha\rangle_6 > \langle\alpha\rangle_{10}$. This difference amounts to 0.3-1.5% for all cases and depends primarily on the crystal lattice type and spacing. Errors of $\geq 1\%$ occurred only for the SC lattice where the target point dependence of α is greatest. Based on this, the target point averaging was done, as a rule, over 6 trajectories.

In the calculation of inclined trajectories ($\varphi \neq 0$) an azimuth averaging of the accommodation coefficient was done for each target point. The precision of this averaging depends on the target point and the angle φ and amounts to ~1.5% for $\varphi = 45°$, on the average. /116

Thus, the selected method of analysis enables one to obtain the parameter values of the individual trajectories with a precision of ~0.1%, and the values of the averaged parameters with a precision of 1.5-2.0%.

4. Some results. By the method, proposed above, the dependence of the average accommodation coefficient on the incident particle energy was determined for the BCC crystal: $\varphi = 0$, $\mu = 0.5$, $s = 0.01$, $a = 0.8$.

The results are presented in Fig. 3. For small w the accommodation coefficient α must be equal to 1 (adsorption). It is difficult to determine precisely the threshold capture energy w_* from the experiments listed. For this case most of the trajectories were found to be "entrapped" when $w = 0.3$ (one of these is shown in Fig. 1). Thus, if one sets $w_* = 0.3$, then we obtain the capture criterion $12\,\mu w_*^2/s = 54$, which is much larger than the value predicted from one-idmensional analyses. From one-dimensional analyses, w_* amounts to ~0.06 for our case. The dependence of α on the crystal type and lattice spacing has been investigated by the same method for the case $w = 1$, $\mu = 0.5$, $s = 0.01$, $\varphi = 0$.

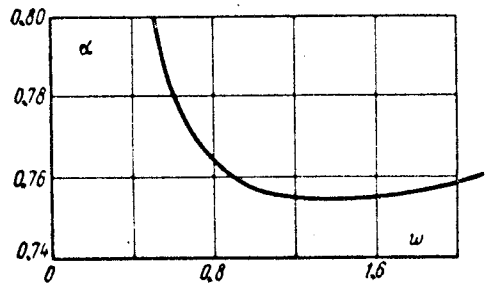

Figure 3. The Dependence of the Accomodation Coefficient on Particle Energy.

The results are presented in Table 2. For the case, designated by the asterisk in the table, the averaging was done over ten trajectories, while in the other cases it was done over six. When an SC lattice with $a = 1.2$ was bombarded, three of the six trajectories, located near the center of the face, were found to be "absorbed" — the particles passed right through the entire atom array. One of these trajectories is shown in Fig. 1, d. For a comparison between the lattices, the results are presented in Fig. 4 as a function of a single parameter—the

TABLE 2. EFFECT OF CRYSTAL TYPE AND LATTICE SPACING

	6-12 Lennard-Jones potential			Morse potential $a\sigma \simeq 7.4$
	SC	FCC	BCC	BCC
0.65	0.7447			
0.8	0.7554	0.6692	0.7576	0.7450
1.0	0.8009	0.7422	0.7975	0.7844
1.2	0.7954	0.7706	0.8266	0.8170
1.5		0.8099 *	0.8841	

specific lattice volume γ, i.e., the volume occupied by one crystal atom. As is known, this quantity is equal to

$$a^3\text{—for SC, } \frac{1}{2}a^3\text{—for BCC and } \frac{1}{4}a^3\text{—for FCC.}$$

A series of calculations have been made to determine the effect of the type of interaction potential and its parameters on the accommodation coefficient obtained.

A comparison was made of two of the most frequently employed potentials: 6-12 Lennard-Jones

$$V(r) = 4\varepsilon\left[r^{-12} - r^{-6}\right]$$

and Morse

$$V(r) = 4\varepsilon\left[e^{-2a\sigma(r-1)} - e^{-a\sigma(r-1)}\right]$$

The depth of the potential well was assumed equal to s for both potentials. The selection of the parameter $a\sigma$ requires an additional condition.

The value of 55/12 is usually assumed for this parameter; this insures that /117 the integral

$$\int_1^\infty V(r)\,dr$$

will be identical for both potentials. This requirement has no physical significance; however, it leads to good agreement of the results at low particle energies when the region $\bar{r} > 1$ plays the major role. In the present calculations the incident particle energy was 50-1000 times greater than the depth of the potential well, and agreement of the results, obtained with these potentials when $a\sigma = 55/12$, was not observed. For such high (compared with ϵ) energies it will be important to match the repulsive portion of the potential ($\bar{r} < 1$); this is achieved if one requires that the distance of nearest approach of particles with energy E_0 be identical. In this case

$$a\sigma = \left(\ln\frac{1+\sqrt{1+E_0/\varepsilon}}{2}\right)\left[1-\left(\frac{2}{1+\sqrt{1+E_0/\varepsilon}}\right)^{1/4}\right]^{-1}$$

The dependence of $a\sigma$ on E_0/ϵ is slight: $a\sigma = 6$ for $E_0/\epsilon = 0$, $a\sigma = 7.4$ for $E_0/\epsilon = 600$ (as mentioned, $E_0/\epsilon = 12\mu w^2/s$).

The results of a calculation of α for such a choice of $a\sigma$ are presented in the last column of Table 2 (the values of the dimensionless parameters are the

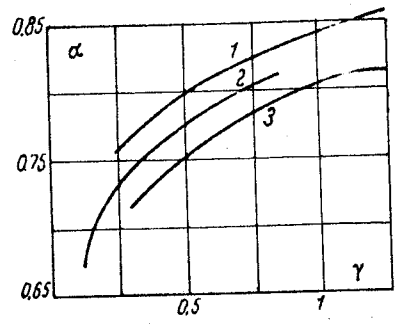

Figure 4. 1—BCC, 2—FCC, 3—SC.

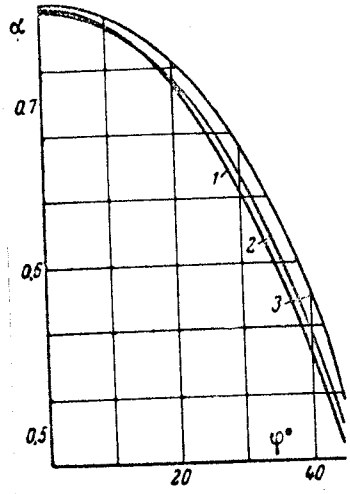

Figure 5. The Dependence of the Accomodation Coefficient on the Incidence Angle: 1) w = 1.0, 2) w = 1.4, 3) w = 2.0.

same as for the rest of the table). The calculated values of the accommodation coefficient are close to the corresponding values obtained by using the 6-12 Lennard-Jones potential. Thus, in our range of parameters both potentials must be acknowledged to be equivalent when $a\sigma$ is suitably chosen. The Lennard-Jones potential was used in all the other calculations.

The effect of the potential well depth on the accommodation coefficient was examined for the case: BCC, w = 1, $\mu = 0.5$, $\varphi = 0$, a = 0.8. The following results are obtained: $a\sigma = 6$ for $E_0/\epsilon = 0$, $a\sigma = 7.4$ for $E_0/\epsilon = 600$

s = 0.01	0.02	0.04
$\langle\alpha\rangle$ = 0.7576	0.7733	0.8077

An increase in the potential well depth leads to some increase in α.

All the results that have been presented above were for the case of normal incidence ($\varphi = 0$) of an atom onto the crystal surface. Shown in Fig. 5 are the results of a calculation of the accommodation coefficent (averaged over azimuth and target points) as a function of the angle φ and the incident particle energy for the case of BCC, $\mu = 0.5$, s = 0.01. As the angle φ is increased, the minimum on the curve α(w) shifts toward lower energies, the reflection approximates the specular mode and the dependence of α on the target point flattens out.

Submitted December 1, 1966

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Translated for the National Aeronautics and Space Administration under Contract NASW-1694 by Scripta Technica, Inc.